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THEORETICAL ALPHA STAR POPULATIONS IN LOADED NUCLEAR EMULSIONS*

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THEORETICAL ALPHA STAR POPULATIONS IN LOADED NUCLEAR EMULSIONS

By F. E. Senftle, T. A. Farley, and L. R. Stieff

ABSTRACT

Calculations have been made of alpha star populations in loaded nuclear emulsions. Equations are presented for calculation of star types for each significant emitter in the three naturally radioactive series. The application of five-branched star populations as a method of quantitative analysis for microgram amounts of thorium is proposed.

INTRODUCTION

Loaded nuclear emulsions have been frequently used to observe the successive alpha particles emitted in the radioactive decay of the uranium and thorium series. When the intermediate members of a series are short-lived, stars consisting of from two to five alpha tracks will be formed. Yagoda (1949) has shown experimentally that the number of stars with a given number of tracks will vary depending on the radioactive series, concentration, and the length of the exposure time. As the half-lives of the decay products of the thorium series are in general shorter than those of the uranium series, for equal amounts the thorium series forms the most five-branched stars. Yagoda has proposed star counting as the basis of a method for thorium analysis.

This theoretical study of alpha star populations in loaded emulsions was undertaken in an effort to find a quantitative method for the analysis of less than microgram amounts of thorium in the

presence of larger amounts of uranium. More recently Frota-Pessoa (1952) has proposed star counting as a method to measure Ra²²⁶ in the presence of thorium. Isaac and Picciotto (1953) have used a similar method for Th²³⁰ (ionium) analysis in deep-sea sediments with considerable success, and Schneider and Matitsch (1952) have used the same technique for actinium analysis.

For these and other applications of loaded nuclear emulsions in the quantitative analysis of radioactive isotopes of the thorium or uranium series, a knowledge of the distribution of various multiplebranched stars for a given concentration and exposure time is desirable. Flamant (1948) has calculated the probability of formation of multiple track stars relative to the number of disintegrations of the parent element in each of the natural radioactive series. However, as the parent elements of each series are decaying at different rates, the results do not conveniently allow comparison of numbers of like stars from the different series. Moreover, whereas the number of disintegrations of each daughter species is equal to the number of disintegrations of the parent member of the series, the number of disintegrations of the original amount of a short-lived daughter nuclide present is not equal to the number of disintegrations of the parent nuclide. Thus, for the shortlived nuclides Flamant's probabilities do not represent the actual star-type distribution in a loaded nuclear emulsion, although for long-lived nuclides they are quite accurate. For these reasons it has been found expedient to compute analytical expressions for each type of star from each of the significantly contributing members of the series as well as summation formulas for the whole series.

METHOD OF CALCULATION

Consider a hypothetical radioactive chain in which each member is in secular equilibrium, and in which the number of atoms, N, of each species is genetically related as follows:

$$N_1\lambda_1 = N_2\lambda_2 = N_3\lambda_3 \dots N_n\lambda_n$$

The transformations of each member of this series during a time, t, can be represented as in figure 1. Of the original number of parent atoms, N₁, some will decay in time t to form ₁N₂ of type-2 atoms. During the same period ₁N₂ type-2 atoms will also be formed by decay of the parent species, N₁, but these will further decay to other daughter products, ₁N₃, ₁N₄, etc., before the end of time t. In a similar manner the number of type-2 atoms, N₂, originally present will decay forming a number of different daughter nuclides, ₂N₃, ₂N₄, etc.

If, for simplicity, we assume that all of the species are alpha emitters then the number of multiple track alpha stars can be readily deduced. Thus the number of single tracks, $_{1}S_{1}$, from type-1 atoms will be $_{1}N_{2}$. Likewise the single tracks, $_{1}S_{2}$, accruing from type-2 atoms will be $_{2}N_{3}$, and the single tracks formed by the series will be

$$I^{S} = \sum_{\substack{j=1 \ j=1}}^{i=n-1} i^{N}_{j} = i^{N}_{2} + i^{N}_{3} + i^{N}_{4} \cdots n^{-1}_{n}$$

In an analogous manner the number of double track stars, from type-1 atoms will be $_1N_3$ and the series equation will be

$$\mathbf{n}^{S} = \sum_{\substack{j=1\\j=3\\i-1}}^{i=n-2} i^{N}j$$

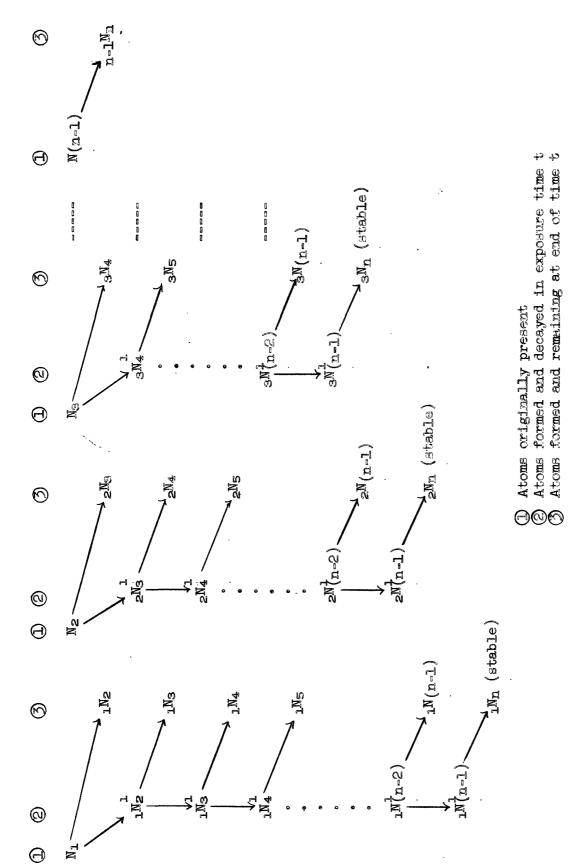


Figure 1. -- Star formation decay scheme

However, the natural radioactive series are made up of beta emitters as well as alpha emitters and the above treatment becomes more complicated because beta particles are not ordinarily observed on alpha sensitive emulsions. Because of these beta emitters a triple alpha track star, for example, may be formed by a series of decays only three of which are alpha decays. Thus, in the thorium series the total number of triple tracks from Ra²²⁴ will not be simply represented by the number of Pb²¹² atoms formed but by the sum of the Pb²¹², Bi²¹² and Po²¹² atoms, i.e. by

Bateman (1910) has given an equation for calculating the number of atoms which decay to and remain at any given member of any series during an exposure period t. In many instances, however, the results obtained by using the conventional form of the equation are subject to large errors. To obviate such errors Flanagan and Senftle (1953) have used an approximation method and have complied tables to facilitate the otherwise laborious calculations. The results and calculations presented here have been obtained from the data in their paper. The formulas in table 1 are based on the amounts of each daughter nuclide which is in equilibrium with one microgram of U²³⁸ or of Th²³². Modes of decay which give rise to an insignificant number of stars during an exposure time up to 500 hours, as compared to other emitters in the series, have been ignored both in the individual and in the summation formulas. Terms in the individual formulas, small in comparison to terms in other individual formulas, have been dropped from the summation formulas.

Table 1.--Individual emitter star formulas for each series

I. Thorium²³² series

A. Single Track Stars

B. Triple Track Stars

C. Quadruple Track Stars

D. Quintuple Track Stars

Table 1. -- Continued,

T mess = 2.154 x 103 e Ma 2 4 - 3.5 x 10e Poste - 2.119 x 103 + 4.125 x 10 3 + 2.384 x 10 11,2 \mathbf{A}_{Ra}^{S} Sa = 1.2 x 10 - 2.6 x 10 $^{-5}$ t + 2.39 x 10 $^{-11}$ t $^{-}$ 1.2 x 10e $^{-}$ Razz4t II. Uranium 238 Series

A. Single Track Stars

18ps10= 2.71 x 102 - 4.48g x 10-4t + 3.711 x 10-10t2 - 7.290 x 10-18t3 - 2.71 x 102 e Bislot LS 4 7.9 x 10 e Abi214^t - 1.08 x 10² e Apb214^t + 2.9 x 10 e J. Razze = 5.887 x 103 e Razzet - 5.887 x 103 e Anzezzt S = 2.1 x 10 e **Pb210t - 2.1 x 10 e **B1214t $T_{U^{238}}^{S} = 1.254 \text{ x } 10^{-2} \text{t}$ $_{\rm I}^{\rm S_{\rm U}234} = 1.25_{\rm S} \times 10^{\rm e}$ $1^{S}_{Th}^{230} = 1.25 \times 10^{-2} t$

B. Double Track Stars

ISpozio= 1.235 x 10-2t - 3.579 x 10 10 t2 + 6.918 x 10 118t3

-181214t - 1.23 x 10 e ... -1.4 x 10e Appg14t ${}^{\rm S}_{\rm Po}$ 218 = 1.1 x 10e ${}^{\rm A}_{\rm Bi}$ 214 t ${}^{\rm A}_{\rm Pb}$ 210 t $\mathbf{\pi}^{S}_{Rn}$ zzz = 5.0 x 10e *Rn zzz + 5e $^{+}$ yezzs + 5e I Shazze = 3e Razzet - 3e Rnzzzt

Table 1. - Continued.

D. Quadruple Track Stars

III. Uranium²³⁵ Series

A. Single Track Stars

 $I_{U^{235}}^{S} = 5.678 \times 10^{-4} t$

B. Quadruple Track Stars

Table 1. -- Continued.

+ 1.245 x 10⁻³⁰t⁵ = 1.48₆ x 10⁻³⁷t³ + 1.52₁ x 10⁻⁴⁴t⁷ + 2 e ^APb²¹¹t JRazza = 5.675 x 10 t - 2.032 x 10 t + 4.853 x 10 17 3 - 8.691 x 10 t

C. Quintuple Track Stars

+ 1.457 x 10 30 t5 = 1.938 x 10 37 t3 + 2.097 x 10 447 = 1.6 x 10 + 1.0 x 10e 4m227 + 6e Razzat , -A_{Ra223}t $\mathbf{x}_{\text{Th}^{227}} = + + + 1.50_4 \times 10^{-3} + 2.01_2 \times 10^{-10} + -7.68_3 \times 10^{-17} + 1.68_7 \times 10^{-23} + \frac{2}{3}$ $\mathbf{x}_{\text{Th}^{227}} = + + 2.01_2 \times 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 10^{-37} + 1$ \mathbf{T}_{Ac} \mathbf{Z}_{Ac} \mathbf{Z}_{C} \mathbf{Z}_{C

Analytical expressions for calculating the number of multiple tracks from the significant members of each of the naturally radioactive series are shown in table 1. The exposure time t is in seconds. The summation formulas for each series are shown in table 2. For the thorium or uranium series comparisons up to an exposure time of 500 hours are shown in figures 2 through 6. When the parent element is in amounts larger or smaller than one microgram, the star population for a given multiple star will be directly proportional to the data in the graphs. The ratio of different star types of a given series, on the other hand, is independent of the amount of parent present.

For chemically separated radioelements loaded into an emulsion, the star population distribution can be computed from the formulas in table 2.

DISCUSSION

Flamant (1948) has calculated the probability of existence of multiple track stars relative to the number of disintegrations of the parent nuclide of the series. For daughter elements with relatively long half-lives the number of disintegrations during the exposure time for the amount originally present will be essentially the same as the number for the parent nuclide of the series. For these cases our calculations are in agreement with Flamant's.

For short half-lived daughter nuclides, however, the number of disintegrations of the atoms originally present during the exposure time is less than the number of disintegrations of the parent nuclide.

Therefore, Flamant's probabilities do not give the actual star distribution in a loaded emulsion for this type of emitter. For example, the star population for Th²²⁸ at 500 hours is compared for the two

Table 2. -- Summation formulas for each series.

+ 3.19 x 10² e Pb^{212t} $^{-1.85}_{9} \times 10^{3} \text{ e}^{-1.85}_{1.0}$ - 1.97s x 103 - 2,424 x 103 e Ra224t = 2 e "Bizlz" + 1.860 x 10³ 2.47 x 10² e Th²³² = 3.46 x 10² e Ra^{228†} = 3.3 x 10 e Th^{228†} - 2.61<u>1 x 10³ e</u> + 2.143 x 103 e $^{-\lambda}$ Ra^{224†} = 3.5 x 10 e $^{-\lambda}$ Pr^{212†} + 4.093 x 10 %: + 2.117 x 103 - 2.51 x 102 e Pb212^t + 4 e B1212^t

S = 2.82 x 102 e Ra²²⁴ - 2.84 x 102 e Pb212^t + 2 c B1212^t

S = 5.46 x 10² e Ra^{228t} - 9.9 x 10 e Pb228^t - 2.42_x x 10³ - 3 $_{\rm I}^{\rm S} = ^{\rm h.07g \times 10^{-3}t} + 2.60_{\rm I} \times 10^{3} \, {\rm e}^{\rm -} {\rm Raszet}^{\rm t}$ $^{-}$ 2 e $^{-}$ $^{\mathrm{A}_{\mathrm{B}1}}$ 212 $^{\mathrm{t}}$

IS = 7.735 x 103 + 4.894 x 10 2t + 1.32 x 10 11t2 - 3.74 x 10 1893 + 5.887 x 103 e Razzet - 5.887 x 103 e Razzet - 1.08 x 102 c Ph214 + 5.8 x 10 e Bi214 + 5.0 x 10 e Ph210t = 7.735 x 103 e Apo^{210t} II. Uranium²³⁸ Series

- 7.0 x 10 e Bi^{214 t} + 5.890 x 10³³ e A^{PD}²¹³t
- 7.0 x 10 e Bi^{214 t} + 5.890 x 10³³ e A^{PD}²¹³t

 $\frac{-\lambda}{12} = -5.9 \mu_3 \times 10^3 + 1.25_5 \times 10^{-2} + 6.2 \mu_8 \times 10^{-12} \mu^2 + 5.9 \mu_3 \times 10^3 \text{ e}^{-1} \text{Ryz22}^{\dagger}$

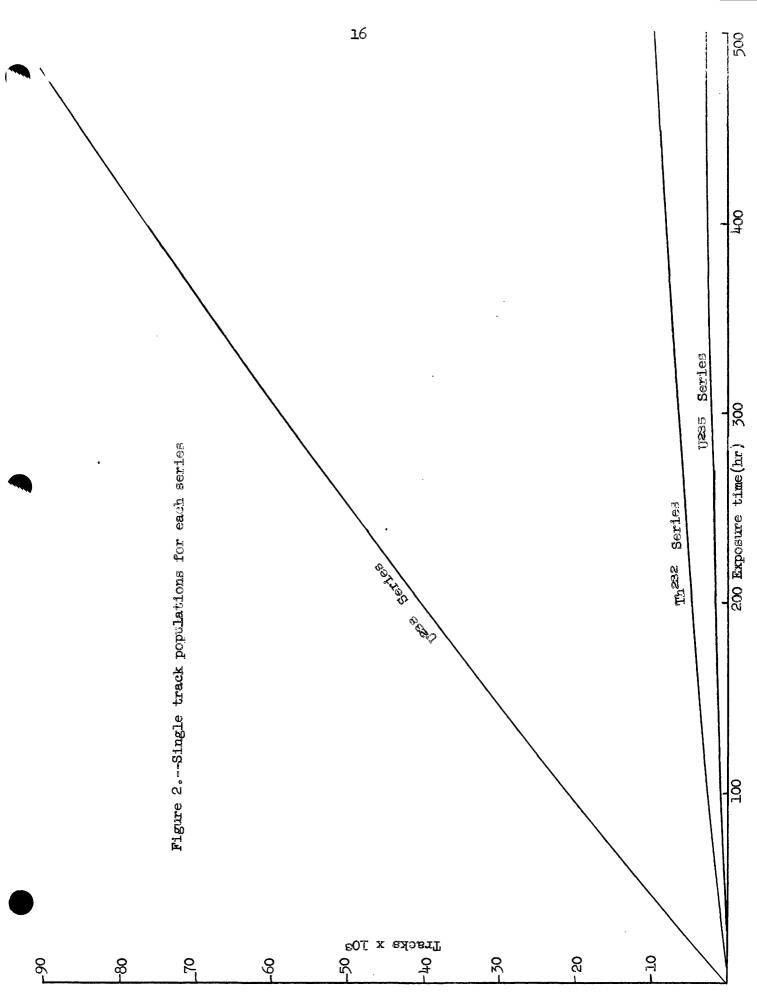
Table 2.--Continued.

III, Uranium 235 Series

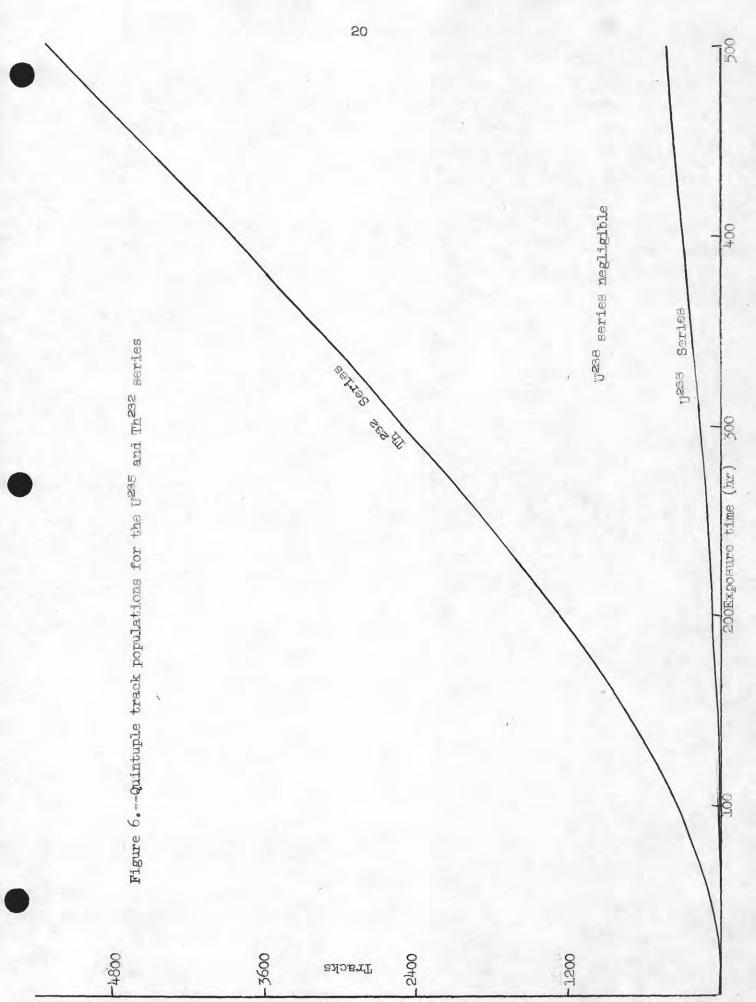
+ 1.229 x 10⁻³ t⁵ - 1.46g x 10⁻³⁷t⁶ + 1.50₂ x 10⁻⁴⁴t⁷ - 1 + 5 e $^{-1}$ In^{227t} + 1.0 x 10 e $^{-1}$ Ac^{227t} - 1.0 x 10 e $^{-1}$ Ra^{223t} - 2 e $^{-1}$ Pb^{211t} $_{\rm T}{
m S}$ = 1.698 x 10⁴³t - 2.012 x 10⁻¹⁰t² + 4.79½ x 10⁻¹⁷t³ - 8.583 x 10⁻²⁴t⁴

 $_{II}$ S = 5.67₅ x 10⁻²⁴t + 1.24₅ x 10⁻³⁷t = 8,69₃ x 10⁻²⁴t + 1.24₅ x 10⁻³⁰t = -1.48₆ x 10⁻³⁷t + 1.52₃ x 10⁻³⁴t - 4 e Razzat + 2 e Appent + 2 e Aczert

 \mathbf{x} = 7.10 x 10⁻⁶t + 2.006 x 10⁻¹⁰t² - 4.79 x 10⁻¹⁷t³+ 8.576 x 10⁻²⁴t⁴ - 1.22₈ x 10⁻³⁰t⁵ + 1.46₅ x 10⁻³⁷t⁶ - 1.49₉ x 10⁻⁴⁴t⁷ - 1.2 x 10 + 1.2 x 10 e



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methods in table 3A. There is essential agreement as the half-life of Th²²⁸ is relatively long (T₂ = 1.90 years). A further comparison is made in table 3B for the short half-life daughter nuclide Ra²²⁴ (T₂ = 3.64 days). Flamant's data are given in percentage of the number of disintegrations of the parent nuclide and they do not add up to 100 percent. The percentage calculated from the formulas in this paper are the percentages which will be observed in a nuclear emulsion provided the star splitting effect due to diffusion is negligible.

Star splitting due to diffusion in the uranium series has been investigated by Frota-Pessôa (1952) and in the thorium series by Eichholz and Flack (1951). In both cases the effect was observed at above freezing temperatures. For analytical work this could probably be minimized by exposing the plates at lower temperatures.

ANALYTICAL APPLICATIONS

The theoretical calculations reported in this paper were undertaken in an effort to develop a method of thorium analysis for less than microgram amounts in the presence of larger amounts of uranium. The quantitative analysis of thorium by determining the abundance of five-branched stars in a loaded nuclear emulsion and the comparison of observed and predicted star populations in loaded emulsions will be presented in a separate paper. In addition, the comparison of observed and predicted star populations may be used to check the half-lives of several members of the uranium and thorium series.

Variations in the star populations with depth in the emulsion may be used to study rates of diffusion in the emulsions of the different

Table 3.--Comparison of Th²²⁸ and Ra²²⁴ star populations determined by Flamant (1948) and by the method presented here.

Track type	Determined by Flamant (1948) (percent)	This paper (percent)
A=	-Th ²²⁸ star population	en grinne diese eine gewegt 3 in der Weite wird die Gewent deut das eine der Geschaft gegen der Weite werden d
Single tracks	25	25
Quadruple tracks	2	3
Quintuple tracks (via Po ²¹² Quintuple tracks (via Tl ²⁰⁸)		72
В	-Ra ²²⁴ star population	
Triple tracks	7	0.3
Quadruple tracks (via Po ²¹²) Quadruple tracks (via Tl ²⁰⁸))	99•6

members of the radioactive series and rates of diffusion of different isotopes of the same radioactive element.

As the uranium analysis can be readily made by conventional chemical methods, the following proposed star counting technique can be used for thorium analysis in low concentrations. To the solution of a sample containing uranium and thorium which has been stripped of its radium and radon but whose specific beta activity is known, add a known amount of Th²³⁴ (as a radio tracer for Th²²⁸). A nuclear emulsion is then loaded by immersion in the resulting solution, dried, and the beta activity per unit area of the emulsion is measured. The volume of solution per unit area taken up by the emulsion is determined by its beta activity. After suitable exposure, the loaded emulsion is developed and the number of five-branched stars per unit area is determined. The number of five-branched stars counted can be corrected by subtracting the five-branched stars produced by the Ra²²³ in the U²³⁵ series as the total uranium concentration is known. For a given exposure the number of micrograms of thorium in the original solution is equal to

Th (micrograms/ml) =
$$\frac{A}{\mathbf{x}^{S}_{Th}} \left(N - \frac{0.00715 \text{ U} \cdot _{\mathbf{x}} S_{U^{235}}}{A} \right)$$

where A is the loading factor in cm²/ml, N is the number of five-branched stars counted, and U is the uranium concentration in micrograms/ml.

If a nuclear emulsion is loaded with a solution of uranium and thorium in equilibrium, the thorium content of the solution can be determined using a method of simultaneous equations similar to that discussed by Schneider and Matitsch (1952). This method is useful for the Th/U ratios in the range from 0.01 to 1.0. If Q₁ and Q₂ are the

track counts of two different exposure times, then the following equations will hold for the thorium and uranium content of a given volume of the emulsion

$$Q_1 = k_1 \text{ Th} + k_2 U$$
, for $t = t_1$
 $Q_2 = k_3 \text{ Th} + k_4 U$, for $t = t_2$

The k constants are the number of five track stars per atom of the parent element. They may be calculated by evaluating the series formula for the respective times and dividing by the number of atoms of parent nuclide in 1 microgram. Thus

$$k_1, k_3 = \frac{x^S_{Th}}{2.59 \times 10^{15}}; k_2, k_4 = \frac{x^S_{U^{235}}}{2.53 \times 10^{15}}$$

The Th/U atomeratio obtained from the above equations must be multiplied by the ratio of atomic weights in order to convert the atom ratio to a weight ratio. Uranium, determined by a conventional analysis, can then be inserted into this ratio to determine the thorium content.

The authors wish to thank F. J. Flanagan and Barbara Schwartz for their assistance in making the calculations. This work is part of a program undertaken by the Geological Survey on behalf of the Division of Raw Materials of the Atomic Energy Commission.

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